Analysis on Photoemission Spectrum of Superconducting FeSe

Rikiya Yoshida[a], Takanori Wakita[b,c], Hiroyuki Okazaki[a,d], Yoshikazu Mizuguchi[c,e], Shunsuke Tsuda[c,f], Yoshihiko Takano[c,e], Hiroyuki Takeya[e], Kazuto Hirata[e], Yukako Kato[g], Takayuki Muro[g], Mario Okawa[h], Kyoko Ishizaka[h], Shik Shin[h], Hisatomo Harima[c,i], Masaaki Hirai[a,b,c], Yuji Muraoka[a,b,c], Takayoshi Yokoya[a,b,c]

[a] *The Graduate School of Natural Science and Technology, Okayama University, Okayama, 700-8530, Japan*
[b] *Research Laboratory for Surface Science (RLSS), Okayama University, Okayama, 700-8530, Japan*
[c] *JST, Transformative Research-Project on Iron Pnictides (TRIP), Chiyoda, 102-0075, Japan*
[d] *JST, CREST, Okayama, 700-8530, Japan*
[e] *National Institute for Material Science, Tsukuba, 305-0047, Japan*
[f] *WPI-MANA-NIMS, Tsukuba, 305-0044, Japan*
[g] *Japan Synchrotron Radiation Research Institute (JASRI)/SPring-8, Sayo, 679-5198, Japan*
[h] *Institute for Solid State Physics, The University of Tokyo, Kashiwa, 277-8581, Japan*
[i] *Department of Physics, Kobe University, Kobe, 657-8501, Japan*

Abstract

In this paper, we present the result of soft x-ray photoemission spectroscopy and its comparison with the density functional calculation. Although local density approximation seems to be a good starting point for describing the electronic structure of FeSe, the simulated spectrum poorly reproduced the structure around E_B = 2 eV. This result suggests the necessity of theoretical treatment beyond local density approximation.

Key words: iron chalcogenide superconductor, FeSe, photoemission spectroscopy, band calculation
PACS: 71.20.-b, 74.25.Jb, 79.60.-i

1. Introduction

FeSe has the simplest crystal structure and the least number of elements among the recently-found iron based superconductors [1] although it still composes of tetrahedrally-bounded (Fe-Se) layers. While there are several reports on the electronic correlations in the family of iron based superconductors [2, 3], one may consider that the simplicity in FeSe yields the easiness of theoretical handling, and studying the electronic structure of FeSe can provide the further insights into the correlation effects. Based on this motivation, we studied the electronic structure of FeSe using soft x-ray photoemission spectroscopy. Since we present the basic information of electronic structure in the previsous work [4], we focus on the comparison of experimental spectra with the result of band calculations.

2. Experimental

We synthesized our FeSe samples using the standard solid state reaction method as described elsewhere [5]. Here we comment on the quality and the stoichiometry of our samples in reference to the validity of our photoemission data. Although we previously assumed the selenium deficiency in our samples [4, 6], there exist the lack of consensus on the stoichiometry of FeSe and the presence of excess iron in the form of oxide or iron metal has been claimed for off-stoichiometric samples [7]. However, while our sample may contain such impurity phases, the effect of such trace impurity phases on our photoemission data should be insignificant. In fact, signal of O 1*s* was not detected in the wide scan.

Our photoemission measurements were performed at two different sites: BL-5 (Okayama University Beamline) of Hiroshima Synchrotron Radiation Center ($h\nu$=140 eV, ΔE=200 meV, @ Room Temp.) and BL27SU of SPring-8 ($h\nu$=1100 eV, ΔE=180 meV, @ 13 K). Samples were fractured *in situ*, and binding energies of the spectra were calibrated in reference to the Fermi edge of a gold film (or molybdenum) evaporated near the sample. The base pressure of main chamber was kept better than 5×10^{-10} torr in these experiments.

In order to obtain some insight on the effect of electronic correlations, we simulated the experimental photoemission spectrum using the calculated electronic partial densities of states (DOS) for FeSe [4], where local density approximation (LDA) is employed. The calcu-

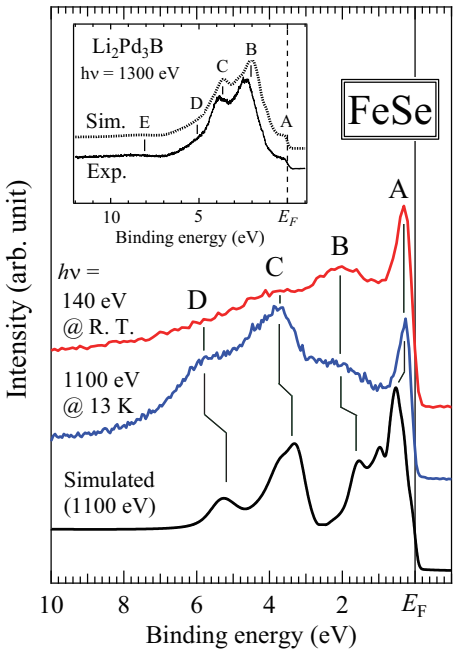

Figure 1: (Color online) Valance band spectrum of FeSe measured at room temperature with photon energy of 140 eV and the one measured at 13 K with soft x-rays of 1100 eV. Comparison of experimental photoemission spectrum with the one simulated from the calculated partial density of states is also presented. (Inset) Similar comparison for the case of Li_2Pd_3B. [9]

lated partial DOS were summed taking photoionization cross sections [8] into account. Then, we convoluted the sum with energy-dependent Lorentzian function (α = 0.10) and Gaussian function (FWHM = 180 meV) to incorporate reasonable life-time broadening and experimental resolution, respectively.

3. Discussion

Figure 1 shows the photoemission spectra taken at two different photon energies together with the simulated spectrum for $h\nu$ = 1100 eV. It is noted here that feature **A** and **B** should correspond to Fe *d*-state; feature **C** shows hybridized state of Fe *d*-state and Se *p*-state; and feature **D** is mainly Se *p*-state. This assignment has been confirmed by the photon-energy dependence of photoemission spectra presented above where feature A and B enhances in $h\nu$ = 140 eV spectrum [4].

One can notice that positions of these four major peaks slightly deviated from the ones of the simulation, from which weak or moderate renormalization of band structures is implied. Moreover, the structure around E_B = 2 eV is poorly reproduced by the simulation. This result is in sharp contrast with the case of Li_2Pd_3B [9] or $Mg_{10}Ir_{19}B_{16}$ [10] where LDA calculation successfully reproduces the experimentally observed spectrum (For Li_2Pd_3B, see the inset). Therefore, our result suggests that we may need to consider the treatment beyond LDA to successfully explain the electronic structure of FeSe. A different approach in treating electronic correlations, such as LDA+DMFT, is encouraged.

Acknowledgements

We would like to thank K. Okada and M. Ichioka for fruitful discussions. We also thank T. Kiss and Y. Nakamura for their help on experiments. For experiments at HiSOR, we acknowledge M. Taniguchi, H. Namatame, and other stuffs there. The synchrotron photoemission measurements at SPring-8 were done under the proposal number of 2008B1581 and 2009A1515. This work is partly supported by a Grant-in-Aid for Scientific Research on Innovative Areas " Heavy Electrons " (No. 20102003) of Ministry of Education, Culture, Sports, Science and Technology, Japan. R. Y. acknowledges the support from JSPS research fellowship.

References

[1] F.-C. Hsu, J.-Y. Luo, K.-W. Yeh, T.-K. Chen, T.-W. Huang, P. M. Wu, Y.-C. Lee, Y.-L. Huang, Y.-Y. Chu, D.-C. Yan, M.-K. Wu, Proc. Natl. Acad. Sci. U.S.A. **105** (2008) 14262.

[2] W. L. Yang, A. P. Sorini, C-C. Chen, B. Moritz, W.-S. Lee, F. Vernay, P. Olalde-Velasco, J. D. Denlinger, B. Delley, J.-H. Chu, J. G. Analytis, I. R. Fisher, Z. A. Ren, J. Yang, W. Lu, Z. X. Zhao, J. van den Brink, Z. Hussain, Z.-X. Shen, T. P. Devereaux, Phys.Rev. B **80** (2009) 014508).

[3] S. L. Skornyakov, A. V. Efremov, N. A. Skorikov, M. A. Korotin, Yu. A. Izyumov, V. I. Anisimov, A. V. Kozhevnikov, D. Vollhardt, arXiv:0906.3218v1

[4] R. Yoshida, T. Wakita, H. Okazaki, Y. Mizuguchi, S. Tsuda, Y. Takano, H. Takeya, K. Hirata, T. Muro, M. Okawa, K. Ishizaka, S. Shin, H. Harima, M. Hirai, Y. Muraoka, T. Yokoya, J. Phys. Soc. Jpn. **78** (2009) 034708.

[5] Y. Mizuguchi, F. Tomioka, S. Tsuda, T. Yamaguchi, Y. Takano, Appl. Phys. Lett. **93** (2008) 152505.

[6] S. Margadonna, Y. Takabayashi, M. T. McDonald, K. Kasperkiewicz, Y. Mizuguchi, Y. Takano, A. N. Fitch, E. Suard, K. Prassides, Chem. Commun. (2008) 5607.

[7] T. M. McQueen, Q. Huang, V. Ksenofontov, C. Felser, Q. Xu, H. Zandbergen, Y. S. Hor, J. Allred, A. J. Williams, D. Qu, J. Checkelsky, N. P. Ong, R. J. Cava, Phys. Rev. B **79** (2009) 014522.

[8] J.J.Yeh, I. Lindau, Atomic Data and Nuclear Tables **32** (1985) 1.

[9] T. Yokoya, T. Muro, I. Hase, H. Takeya, K. Hirata, K. Togano: Phys. Rev. B **71** (2005) 092507.

[10] R. Yoshida, H. Okazaki, K. Iwai, K. Noami, T. Muro, M. Okawa, K. Ishizaka, S. Shin, Z. Li, J. L. Luo, G.-q. Zheng, T. Oguchi, M. Hirai, Y. Muraoka, T. Yokoya, J. Phys. Soc. Jpn. **78** (2009), 034705.